PHYSICS STUDY FOR A LEU MO TARGET IRRADIATION AT HANARO

B.C. Lee, C.S. Lee and H. Kim
Research Reactor Analysis Department
KAERI, P.O. Box 105, Yuseong, Daejeon, 305-600, KOREA

ABSTRACT

Studies on the target irradiation for fission Mo production at HANARO have been steadily progressed. Low enriched uranium foil, which is the most promising LEU target for fission Mo production and is now under development here, is adopted as a target. Physics concerns are reviewed in some detail with an assumption that the LEU foil target is being irradiated at the OR irradiation hole in HANARO. The LEU foil target geometry is decided considering the OR hole size and the domestic demand for Mo-99 activity. It is an annulus type and its thickness is 100μm. Reactivity change due to target loading is examined to confirm the reactor safety. It is estimated to be less than 0.8mk with a standard deviation of 0.24mk. Even the irradiation of the two targets gives only 1.6mk of a reactivity change including an uncertainty. This is much lower than the safety limit of an experiment. The target will be loaded and unloaded during the reactor operation. Considering the small reactivity effect of the target, the irradiation activities of the target will not cause severe reactivity induced accidents. The thermal neutron fluxes at the vertical irradiation holes and horizontal beam tubes in the reflector should not be perturbed much due to the target irradiation. Most of the fluxes are maintained within a 2% fluctuation. The calculated powers at one target are in the range of 30~36kW. This power can produce a specific activity of 45~54 Ci Mo-99/gU at the end of irradiation.

1. Introduction

Mo-99 is one of the most important radioisotopes in nuclear medicine. Even though the demand for Mo-99 is small, it is indispensable in cancer diagnosis. Mo-99 has a relatively short half-life of 66.7hrs. Thus, a continuous and constant supply is important. Currently, the domestic demand of Mo-99 has been totally fulfilled by imported ones. Since HANARO's first operation in 1995, a study on producing Mo-99 with a highly enriched uranium target has been conducted to fully utilize HANARO for several years[1,2]. Various target materials and geometries proper for the irradiation hole were studied to maximize fission Mo production. In addition, the chemical process for Mo extraction and the manufacture of the Cintichem type target were actively tested. However, use of HEU became difficult because of nuclear nonproliferation.
In the meantime, we launched a program for the development of fabrication technology for uranium foil which was considered as the most promising LEU target for fission Mo production, and succeeded in developing a trial product of the uranium foil of 100~200μm in thickness[3]. With the good R&D infrastructure through the HEU study and the LEU target fabrication technology, the program for fission Mo production was redirected to use the LEU target.
It is expected that there will be a gradual increase of the Mo-99 demand in the domestic fields according to the economic growth. An increase of the terror threat for air transportation changes the situation for a constant supply of Mo-99. A research reactor itself needs a periodic shutdown for maintenance. These

environments require a regional organization for a backup supply system for radioisotopes among the neighboring countries. It requires the production of the main radioisotopes such as Mo-99.
Physics concerns for irradiating the fission Mo target are the reactor safety according to the loading of the fissionable material into the reactor core, target integrity during the irradiation, etc. The reactivity, fission power and perturbation effect for the other irradiation sites are evaluated in this paper.

2. Target Design and Analysis

2.1 Target design

Two types of UO_2 Cintichem and U metal foil were studied as a LEU target[4,5]. Intensive analyses of various target materials and geometries were previously performed through the HEU study. We decided to use the U metal foil as a target since it is the best substitute for the HEU target considering the amount of radioactive waste during the chemical process after the target irradiation.
HANARO has many irradiation sites in the core and reflector regions as shown in Figure 1. For the fission Mo target irradiation, the OR sites near the core are proper considering the flux level and availability. In the analysis, OR5 is chosen for the target irradiation because the in-chimney bracket to fix the on-power irradiation facility is available. The thermal neutron fluxes at OR5 are about 2×10^{14} n/cm^2-s. The saturated Mo-99 activity from fission is 51.5Ci/kW. Assuming 5 days' irradiation, about 20~25gm U of 20% LEU should be irradiated in order to produce about 1200Ci of Mo-99 at the end of the irradiation for the domestic demand and a reserve. The inner radius of the OR is 30mm. It is preferable to irradiate the target at the axial position of the peak neutron flux in order to maximize the Mo-99 production. A shorter length of the target is better because of a higher average neutron flux. An annulus type of the U foil was chosen as a LEU fission Mo target and was 100μm in thickness and 100mm in length. The U foil was coated with Ni and then clad with aluminum. The target cross section is shown in Figure 2.

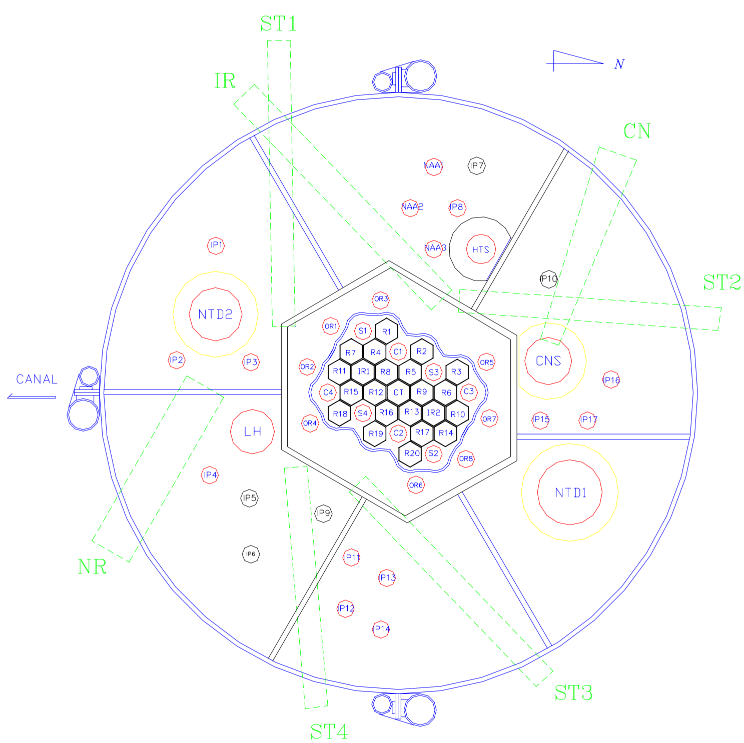

Fig. 1. HANARO core plan view

2.2 Reactivity

The reactivity and power of the target and neutron flux at the irradiation sites are calculated for an equilibrium core using MCNP. The number densities of the fission products for the burned fuel in the MCNP input are calculated using HANAFMS (HANARO Fuel Management System) consisting of VENTURE and WIMS-KAERI[6]. The assembly-wise axial burnup distribution of each fuel assembly is calculated from HANAFMS, and the burnup dependent nuclide number densities are from the WIMS-KAERI lattice calculation. It includes the 20 main fission products for an efficient and simple calculation[7]. The CARs are located 450mm from the fuel bottom.

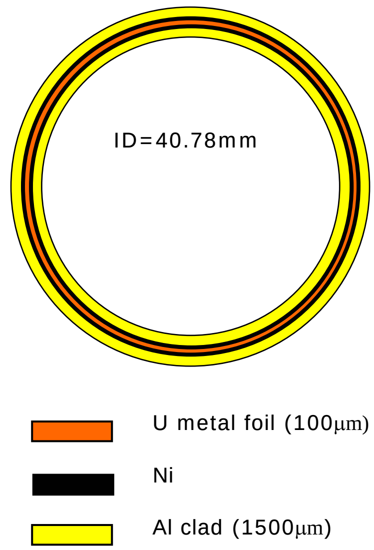

Fig. 2. U metal foil target

The target irradiation site is OR5 but the effect of irradiating the target at OR3 is also checked. The U foil will be fabricated with some surface roughness. Considering a thin thickness of around 100μm, the surface roughness affects the uranium mass of the target and then the Mo-99 activity. The surface roughness will be controlled to within an acceptable limit in the fabrication process. To evaluate the effect of the roughness, the target foil thickness was varied from 75μm to 125μm in the analysis. It is also calculated for the case of the two targets irradiation at the same time.
The reactivity change due to target loading is much lower than the HANARO safety limit as expected. When the two targets are loaded, it is only 1.15mk with 0.33mk at 1 σ.

Table 1. Reactivity change due to fission Mo target loading

Case	Hole	Foil thickness (μm)	k-effective	Reactivity (mk)
A	OR5	-	0.99525(0.00022)*	-
B	OR5	100	0.99578(0.00024)	0.535
C	OR3	100	0.99597(0.00022)	0.726
D-1	OR5	75	0.99603(0.00024)	0.787
D-2	OR5	80	0.99553(0.00021)	0.283
D-3	OR5	90	0.99583(0.00024)	0.585
D-4	OR5	110	0.99590(0.00022)	0.686
D-5	OR5	120	0.99599(0.00023)	0.747
D-6	OR5	125	0.99606(0.00022)	0.817
E**	OR5	100	0.99639(0.00022)	1.150

* fractional standard deviation
** two targets are irradiated

Since the target will be loaded during reactor operation, the target may perturb the core condition. The target loading speed should be decided in order that the reactivity insertion rate into the reactor is much lower than the limit to guarantee reactor safety. In consideration of a small reactivity as above, the target loading speed can be decided to satisfy the limit of 0.125mk/s. If the target is loaded for 100 sec into the core region of 50cm, for instance, the loading speed is 0.5cm/s and the reactivity insertion rate can be conservatively controlled within 0.02mk/s.

2.3 Perturbation effect on other irradiation sites

There are many utilization activities such as neutron beam research, NTD-Si irradiation, radioisotope production, etc. at HANARO. These activities are expected to increase continuously. It is very important to maintain a stable neutron flux at the irradiation sites including the beam tube. Thus, the neutron fluxes at the irradiation sites should not be perturbed much due to the fission Mo target loading. The perturbation effect on the neutron flux is calculated.

The changes of the axial average thermal neutron fluxes at the irradiation sites are tabulated in Table 2. In most cases, the thermal neutron fluxes are perturbed within 2%. At the irradiation sites near the fission Mo target, the difference is a little higher but within 5%.

Table 2. Neutron flux perturbation at vertical irradiation sites due to fission Mo target loading

Irradiation Site	Average thermal neutron flux (n/cm^2-s)	Difference of average thermal neutron flux (%)		
	Al dummy fuel at OR5	Target at OR5	Target at OR3	2 targets at OR5
IP1	4.99E+13 (0.0064)*	1.15	2.10	-0.36
IP2	4.66E+13 (0.0062)	-1.32	0.07	-2.08
IP3	9.08E+13 (0.0037)	-1.16	0.41	-1.76
IP4	6.48E+13 (0.0054)	-1.80	1.13	-2.07
IP5	5.53E+13 (0.0046)	0.37	1.71	-0.14
IP6	5.66E+13 (0.0059)	-0.69	0.21	-1.22
IP7	2.75E+13 (0.0080)	-1.23	2.57	0.87
IP8	5.28E+13 (0.0059)	-0.64	0.98	0.41
IP9	1.52E+14 (0.0037)	0.41	1.08	-0.60
IP10	4.38E+13 (0.0050)	4.08	1.76	4.37
IP11	7.03E+13 (0.0041)	-0.14	1.17	-0.51
IP12	5.52E+13 (0.0059)	-0.30	-0.44	0.15
IP13	9.92E+13 (0.0046)	-0.42	-0.89	-0.79
IP14	4.90E+13 (0.0063)	0.99	-0.15	0.22
IP15	9.70E+13 (0.0036)	1.46	-0.43	2.72
IP16	4.92E+13 (0.0062)	2.14	-0.53	4.42
IP17	3.99E+13 (0.0052)	2.24	0.03	4.26
NAA1	2.39E+13 (0.0088)	-1.37	2.38	-0.62
NAA3	8.88E+13 (0.0052)	-0.05	1.34	0.62
HTS	4.06E+13 (0.0063)	2.04	1.71	0.65
LH	1.16E+14 (0.0029)	-0.78	0.35	-1.19
CNS	8.21E+13 (0.0031)	2.74	0.10	3.57
NTD2	4.52E+13 (0.0035)	-0.06	1.81	-0.69

* fractional standard deviation

The thermal neutron fluxes at the beam tube nose are summarized in Table 3. The differences of the thermal neutron fluxes due to target loading are all within 2%.

Table 3. Thermal neutron flux perturbation at beam tube nose due to fission Mo target loading

Beam tube nose	Thermal neutron flux (n/cm^2-s)	Difference of thermal neutron flux (%)		
	Al dummy fuel at OR5	Target at OR5	Target at OR3	2 targets at OR5
IR	2.57E+14 (0.0055)*	0.99	0.75	0.30
NR	6.90E+13 (0.0085)	-1.00	0.63	-0.53
ST1	1.81E+14 (0.0067)	-1.13	1.03	-1.41
ST2	2.04E+14 (0.0062)	-0.12	0.08	0.61
ST3	2.50E+14 (0.0056)	-0.24	0.21	-0.82
ST4	1.90E+14 (0.0063)	-0.87	-0.49	-2.04

* fractional standard deviation

From the above results, it is concluded that the fission Mo target loading at OR3 or OR5 does not perturb the thermal neutron flux at the other irradiation sites. It means that the target can be loaded regardless of the other irradiation activities. However, the effect should be confirmed in detail before the actual irradiation of the target.

3. Target Power and Mo Activity

The fission power at the target for each case is listed in Table 4. It is about 32kW at one target. This power generates about 50Ci Mo-99/gU at the end of irradiation. If the target is irradiated at OR3, the power and Mo-99 activity are 14% higher than those at OR5.

Table 4. Target power and Mo-99 activity

Case	Hole	Foil thickness (μm)	U mass (g)	Target power (kW)	Mo-99 activity at the end of irradiation (Ci Mo-99/gU)
B	OR5	100	24.4	32.7 (0.0077)*	49.1
C	OR3	100	24.4	37.3 (0.0073)	56.1
D-1	OR5	75	18.3	25.9 (0.0079)	38.9
D-2	OR5	80	19.5	27.3 (0.0078)	41.0
D-3	OR5	90	22.0	30.1 (0.0078)	45.2
D-4	OR5	110	26.8	35.6 (0.0076)	53.5
D-5	OR5	120	29.3	37.3 (0.0076)	56.1
D-6	OR5	125	30.5	38.8 (0.0076)	58.3
E	OR5	100	24.4 24.4	32.2 (0.0077) 27.4 (0.0084)	48.4, 41.2

* fractional standard deviation

The domestic demand for Mo-99 activity can be produced from one target. It is expected that there will be a 10% increase for the demand annually. This growth may be covered by irradiating two targets or increasing the foil thickness. A backup supply to neighboring countries may also be possible.

4. Conclusion

An annulus type of uranium metal foil was chosen as a fission Mo target at HANARO rather than the UO_2 Cintichem type target. The uranium foil of about 100μm in thickness is being developed at KAERI. Using this foil, the study on the production of Mo-99 at HANARO has been continued. The U foil of 100μm in thickness and 100mm in length was selected considering the size of the irradiation site, OR.
The reactivity effect due to target loading is estimated to be much smaller than the limit. Also, the target can be loaded into the core within the limit of the reactivity insertion rate. Thus, the operating reactor can be kept safe regardless of the fission Mo target irradiation. The target power was about 33kW and the Mo-99 activity at the end of 5days' irradiation was 49Ci Mo-99/gU. This activity covers the domestic demand. The annual increase and backup to neighboring countries can be supplied by increasing the number of the targets and the U foil thickness.
Some accident scenarios are considered such as dropping the target during loading/unloading and irradiation, target melting at a local point, etc. Most of the accident scenarios are expected to result in a negligible impact due to the small reactivity, but some cases will be analyzed in detail to confirm the reactor safety.

Acknowledgements

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5. References

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